Characteristics of Type I PSCs Derived from POAM Observations

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Abstract. The characteristics of POAM 3 observations of Type I Arctic PSCs from 1998 to 2003 are studied using a scheme that discriminates Type Ia from Ib PSCs. The PSCs observed in these years are studied simultaneously by aligning the day in each year when the temperature associated with a POAM observation first reaches T_{NAT}. It is observed that PSC formation occurs within days of the minimum observation temperature reaching T_{NAT} and that the majority of these first PSCs are Type Ia. Our observations support the hypothesis that heterogeneous freezing contributes at least in part to the freezing of solid phase PSCs.

Introduction

Type I Polar Stratospheric Clouds (PSCs), are generally classified into two groups, Ia and Ib. Type Ib clouds are thought to be composed of small liquid particles, probably ternary solutions of $H_2SO_4/HNO_3/H_2O$. Type Ia clouds are consistent with a small number of larger solid particles that are assumed to be composed of hydrates of nitric acid. Both kinds of Type I PSCs provide surfaces for heterogeneous reactions that produce active forms of chlorine, such as ClO, and that react rapidly to destroy ozone. The larger, solid phase Type Ia PSC particles can permanently remove nitric acid from the stratosphere by sedimentation. This process, called denitrification, allows chlorine to remain in its active state, increasing ozone loss. One study found that as much as 30% of the ozone loss at 20 km during the 1999-2000 Arctic winter was attributable to denitrification by PSCs. [*Drdla et al.*, 2003]

Despite years of study, the formation mechanisms governing Type Ia PSC formation remain uncertain [*Tolbert and Toon*, 2001]. Models of homogeneous freezing predict that air parcels remain at temperatures as much as 5 degrees below T_{NAT} for days before solid-phase particles form and grow large enough to be observed [*Carslaw et al.*, 2002; *Fahey et al.*, 2001]. Accordingly, one would not expect to find any Ia PSCs forming unless the air parcel spent days below $\Delta T = -5\ K$. The heterogeneous theory relies on the existence of 'special nuclei' [*Tolbert and Toon*, 2001] that act as nucleation sites for freezing of nitric acid shortly after T reaches T_{NAT}. The purpose of this paper is to examine PSC characteristics over the last several Arctic winters using POAM satellite observations and taking advantage of a method of discriminating Type Ia and Ib PSCs [*Strawa et al.*, 2002] to shed light on formation mechanisms.

The PSC Discrimination Method

Discriminating between Type Ia and Ib PSCs using satellite occultation measurements is significant because it allows the study of the evolution and effect of PSCs using the more continuous temporal and larger areal coverage that can be obtained from satellites. A method for discriminating Type Ia and Ib PSCs from POAM satellite occultation measurements of aerosol extinction coefficient has been developed [*Strawa et al.*, 2002]. The method takes advantage of the fact that the size and number density of Type Ia and Ib clouds differ greatly. This is reflected in the cloud extinction coefficient and the wavelength dependence (as represented by the Ångström coefficient) measured by occultation satellites. We represent the multispectral dependency of these clouds by a quadratic equation. The linear term in the quadratic fit, a_1, is a modified Ångström coefficient that represents the linear relationship between the observations and the wavelength.

The observations are separated into 4 potential temperature bins, from 400 K to 600 K. As the background sulfate aerosols grow into PSCs, their extinction increases, and PSC observations of Type Ia and Ib bifurcate with differing wavelength dependence. Typically, Ib clouds have higher extinctions and greater wavelength dependencies, whereas Ia PSCs have lower extinctions and less wavelength dependence. Thresholds used for discriminating PSC type which are based on POAM PSC observations and statistics derived from simulations. The method has been validated by applying several statistical tests to the results and by using DIAL and OLEX lidar observations made during SOLVE/THESEO 2000. [*Strawa et al.*, 2002]

Characteristics of the Arctic Winters

The typical Artic winter is characterized by a general cooling and the formation of a polar vortex in November. This study is focused on early PSC formation at POAM latitudes, 60N to 70N, and over winters since 1998. Due to the annual variation of Arctic meteorology, each winter has a very distinctive temperature history. In an attempt to find some common ground between these diverse years to facilitate the study of Type I PSC formation mechanisms, we have shifted each year so that the day when the minimum observed temperature first reaches is aligned.

The Early Formation of PSCs

The aim of this study is to identify PSC formation and evolution characteristics common to all of the Arctic winters since 1998. Each winter was divided into 14-day periods, with the period corresponding to the first occurrence of $\Delta T=0\ K$ designated as period 0. PSC observations were classified into Type Ia, Ib, and mixed clouds using our method. The data, presented in this way show several relevant features.

First, over the course of the winter 52% of the PSCs observed are Type Ia PSCs and only 28% are Ib's. In the first 14-days after T decreased below T_{NAT}, more Type Ia PSCs are observed than Ib and mixed PSCs. Type Ia clouds make up about 56%, Ib clouds 35%, and mixed

clouds 9% of the observations in this period. This phenomena occurs in all years that had significant PSCs observations and indicates that Ia growth times are consistently short and that the formation of Ia clouds occurs shortly after the temperature drops below T_{NAT}.

Typically, the minimum vortex temperature falls below T_{NAT} several days before POAM observed a minimum temperature below T_{NAT} and it is possible that PSCs are formed in a colder part of the vortex and are transported into POAM latitudes. The Goddard Space Flight Center's fast trajectory model [*Schoeberl et al.*, 2000] using UKMO analysis was used to obtain ten-day back trajectories for all PSCs observed in Period 0. Distributions were constructed to look at the total number of days an air parcel associated with a PSC spent below ΔT = -3 K. As expected, the Ib PSCs form quickly since they equilibrate with their environment in a matter of hours. The Ia's, on the other hand, take days to equilibrate and are likely in non-equilibrium with their environment. Still, nearly half of the observed PSCs in Period 0 have not spent any time at ΔT < -3 K, and less than 10% spent more than 3 days below ΔT < -3 K. Thus the appearance of so many Ia's in period 0 is indicative of a formation mechanism that acts mush faster than the homogeneous freezing process.

Large Ia particles can sediment rapidly, on the order of 0.5 km per day. It is possible that an observed Ia PSC actually formed at a higher altitude and sedimented into the layer of observation. To explore this possibility, 10-day back trajectories for all Ia PSC observations in period 0 were calculated at an altitude 2 km higher than the observations and shows that about 75 % of these air parcels spent no time below ΔT = -3 K and only 2 parcels spent more than 3 days below ΔT = -3 K. This finding is supported by other studies that have concluded that freezing takes place above the ice frost point [*Drdla et al.*, 2002; *Tabazadeh and al.*, 2001; *Toon and al.*, 2000].

Conclusions

The formation and evolution of Type I PSCs have been studied by characterizing POAM Arctic PSC observations from 1998 to 2003. The observations of all of the years have been grouped together by shifting each year to align the day when the observed POAM minimum temperature first reaches T_{NAT}. PSC statistics for each of the shifted years are binned into 14-day periods. A scheme to discriminate Type Ia from Ib PSCs is employed in the study. The kind of statistical approach used here is appropriate for the study of formation and evolution of PSCs because the effects of random uncertainties in the observations are reduced.

It was observed that Type Ia PSCs formed within days of the minimum temperature associated with a POAM measurement reaching T_{NAT}. Within the first 14-day period after the minimum temperature reached T_{NAT}, period 0, 56% of the PSCs were Ia's compared with 35 % Ib's. A study of ten-day back trajectories on the PSCs observed in period 0 revealed that 44% of the Type Ia PSCs observed spent no time below $\Delta T = T_{obs} - T_{NAT} = -3$ K, and 93% spent 3 days or less at this temperature. Further, a study of ten-day back trajectories for air parcels originating 2 km above the observed PSCs in period 0 revealed that 67% spend no time at ΔT < -3 K, and 96% spent 3 days or less at this temperature. The implication is that Type Ia PSCs

form quickly in response to the temperature falling below T_{NAT} and need not spend days at temperatures of ΔT < -5 K. Homogeneous models that require a parcel remain below ΔT = -5 K for extended periods of time cannot explain this observed behavior and the observations support the hypothesis that heterogeneous freezing contributes at least in part to the freezing of solid phase PSCs.

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